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Polarization of light scattered into second harmonic by free electrons

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The classical theory of the scattering of light into harmonics by free electrons has been given by one of us (Vachaspati 1962, 1963). If the incident light is plane polarized, the scattered light is also plane polarized. We calculate there the dependence of the angle of polarization of the scattered light as a function of the polarization angle of the incident light and the angle of scattering. The results are discussed in the last section.

Definitions

We call the direction of the electric field as the direction of polarization. To choose appropriate coordinate systems for defining the polarization angles, we take a plane (the plane of the paper) containing the incident light and the observed light with unit vectors $\boldsymbol{n}_0$ and $\boldsymbol{n}$ respectively. Introduce a unit vector $\boldsymbol{\alpha}_1$ perpendicular to $\boldsymbol{n}$ in the plane of the paper and another unit vector $\boldsymbol{\alpha}_2$ perpendicular to the plane of the paper such that the unit vector $(\boldsymbol{\alpha}_1, \boldsymbol{\alpha}_2, \boldsymbol{n})$ form a right handed coordinate system. Explicitly,

$$\boldsymbol{\alpha}_1 = \frac{\boldsymbol{n}_0-(\boldsymbol{n}_0\cdot\boldsymbol{n})\boldsymbol{n}}{\sqrt{1-(\boldsymbol{n}_0\cdot\boldsymbol{n})^2}}$$

$$\boldsymbol{\alpha}_2 = \frac{\boldsymbol{n}\times\boldsymbol{n}_0}{\sqrt{1-(\boldsymbol{n}_0\cdot\boldsymbol{n})^2}} \tag{1}$$

The angle which the electric vector of the scattered light makes with $\boldsymbol{\alpha}_1$ is called the angle of polarization of the scattered light and is denoted by ϕ.

A similar coordinate system is defined for the incident light. The unit vectors $\boldsymbol{\beta}_1$, $\boldsymbol{\beta}_2$ and $\boldsymbol{n}_0$ form a right handed system; explicity,

$$\boldsymbol{\beta}_1 = \frac{\boldsymbol{n}-(\boldsymbol{n}_0\cdot\boldsymbol{n})\boldsymbol{n}_0}{\sqrt{1-(\boldsymbol{n}_0\cdot\boldsymbol{n})^2}}$$

$$\boldsymbol{\beta}_2 = \frac{\boldsymbol{n}\times\boldsymbol{n}_0}{\sqrt{1-(\boldsymbol{n}_0\cdot\boldsymbol{n})^2}} = \boldsymbol{\alpha}_2 \tag{2}$$

Notice the minus sign in $\boldsymbol{\beta}_2$; it has been inserted so that the directions $\boldsymbol{\alpha}_1$ and $\boldsymbol{\beta}_1$ coincide for forward scattering. The angle which the electric vector of the incident light makes with $\boldsymbol{\beta}_1$ is called the polarization angle of the incident light and is denoted by ϕ_0.

The angle of scattering is denoted by θ.

Electric field

The incident beam is taken as plane polarized; its electric field is

$$\boldsymbol{E} = E_0\boldsymbol{e}_0 \cos(k_0(x_0-\boldsymbol{n}_0.\boldsymbol{x})) \tag{3}$$

$$(\boldsymbol{e}_0\, \boldsymbol{\beta}_1) = \cos\phi_0; \; (\boldsymbol{e}_0.\boldsymbol{\beta}_2) = \sin\phi_0. \tag{4}$$

The electric field of the scattered light can be obtained from the expressions given in Vachaspati, (*Phys. Rev.* 1963); it is

$$\boldsymbol{E}^{Scatt} = \frac{e}{r}\{\boldsymbol{M}-(\boldsymbol{n}.\boldsymbol{M})\boldsymbol{n}\}.$$

For the second harmonic light, $\boldsymbol{M}$ can be replaced by

$$\boldsymbol{N}^{(2)} \sin 2\psi_0$$

where

$$\boldsymbol{N}^{(2)} = C^{(2)}\boldsymbol{e}_0+\boldsymbol{D}^{(2)}\boldsymbol{n}_0$$

$$C^{(2)} = -2k_0 q\cos\alpha, \quad D^{(2)} = -\tfrac{1}{2}k_0 q$$

$$q = \frac{e^2E_0^2}{m^2k_0^2}; \quad \cos\alpha = (\boldsymbol{n}.\boldsymbol{e}_0); \quad \psi_0 = k_0(x_0-|\boldsymbol{x}|)$$

so that

$$\boldsymbol{E}^{Scatt} = \frac{e}{r}\,[C^{(2)}\boldsymbol{e}_0+D^{(2)}\boldsymbol{n}_0-\{C^{(2)}(\boldsymbol{n}.\boldsymbol{e}_0)+D^{(2)}(\boldsymbol{n}.\boldsymbol{n}_0)\}\boldsymbol{n}]\sin 2\psi_0 \tag{5}$$

Notice that the electric vector, $\boldsymbol{E}^{Scatt}$, is transverse to $\boldsymbol{n}$:

$$\boldsymbol{E}^{Scatt}\,\boldsymbol{n} = 0$$

as it should be.

Polarization

We resolve $\boldsymbol{e}_0$ and $\boldsymbol{n}_0$ along the three axes provided by $\boldsymbol{\alpha}_1$, $\boldsymbol{\alpha}_2$ and $\boldsymbol{n}$. The result

$$\boldsymbol{E}^{Scatt} = \frac{e}{r}\mathrm{A}(\cos\phi\boldsymbol{\alpha}_1+\sin\phi\boldsymbol{\alpha}_2)\sin 2\psi_0, \tag{6}$$

where

$$\mathrm{A}\cos\phi = C^{(2)}\cos\theta\cos\phi_0+D^{(2)}\sin\theta, \tag{7a}$$

$$\mathrm{A}\sin\phi = C^{(2)}\sin\phi_0. \tag{7b}$$

If we keep our polarimeter so as to receive the incident light, we can find ϕ_0 by measuring the angle which the electric vector makes with $\boldsymbol{\beta}_1$ in the plane $(\boldsymbol{\beta}_1, \boldsymbol{\beta}_2)$; the angle ϕ is measured when the instrument is receiving the scattered light and the angle is in the $(\boldsymbol{\alpha}_1, \boldsymbol{\alpha}_2)$ plane.

When the values of $C^{(2)}$ and $D^{(2)}$ given above are used, we find

$$\cos\phi = \frac{2\cos\theta\cos^2\phi_0 -}{\sqrt{(2\cos\theta\cos^2\phi_0-\frac{1}{2})^2+\sin^2 2\phi_0}}, \tag{8a}$$

$$\sin\phi = \frac{\sin 2\phi_0}{\sqrt{(2\cos\theta\cos^2\phi_0-\frac{1}{2})^2+\sin^2 2\phi_0}}. \tag{8b}$$

Discussion

From (8a, b), we notice that

(1) If $\phi_0 = 0$ then $\phi = 0$ for all θ. This means that, if the incident light is polarized in the plane of the paper, the scattered light is also polarized in the plane of the paper, no matter what the angle of scattering is (except for $\theta = 76°$, in which case the light is perpendicularly polarized, see (3) below.).

(2) If $\phi_0 = \pi/2$, then $\phi = \pi$ for all θ. This means that, if the incident light is polarized perpendicular to the plane of the paper, the scattered light is polarized in the plane of the paper, no matter what the angle of scattering is.

(3) If the incident light polarization is such that

$$\cos\phi_0 = \tfrac{1}{2}\sqrt{\sec\theta}$$

then $\phi = \pi/2$, i.e., the scattered light is polarized perpendicular to the plane of the paper. This relation is possible only if

$$\tfrac{1}{2}\sec\theta < 1 \qquad \text{i.e. } 0 \leqslant \theta \leqslant \cos^{-1} 1/4.$$

Since $\cos^{-1} 1/4$ 76°, this means

$$0 < \theta \lesssim 76°.$$

For angles of scattering greater than 76°, the scattered light cannot be polarized perpendicular to the plane of the paper.

For angles of scattering, θ, less than 76°, the polarization of scattered light will be perpendicular to the plane of the paper if ϕ_0 is chosen as follows :

For $\theta = 0°$, choose $\phi_0 = 60°$
$\theta = 30°$, $\phi_0 \simeq 57{\cdot}5°$
$\theta = 45°$, $\phi_0 \simeq 53{\cdot}5°$
$\theta = 60°$, $y_0 = 45°$
$\theta = 76°$, $y_0 \simeq 0e.$

(4[The dependence of the angle of polarization of the scattered light on the polarization angle of the incident light and the angle of scattering can be plotted and is given in the accompanying figure.